

# A Nano-Quantum Photonic Model for Justification of Dispersion in Single Crystal Film of NPP

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## Abstract

In this paper, we present a nano-quantum photonic model for justification of normal dispersion in a thin crystal film of NPP. In this method, we assume a laser beam consists of a flow of energetic particles. By precise analyzing of photon interaction with  $\pi$ -electron system of benzene ring in NPP crystal, we will attain refractive index (RI) in any wavelength and compare the results with experimental data.

*Keywords:*  $\pi$ -electron system, photon-electron interaction, dispersion, NPP.

## 1 Introduction

Organic optical materials like MNA, NPP, MAP have a high figure of merit in optical properties in comparison with inorganic optical materials such as LiNbO<sub>3</sub>, ADP, KDP and GaP [1]. Additionally N-(4-nitrophenyl)-L-prolinol (NPP) after MNA has the highest figure of merit between organic nonlinear optical materials [2]. Thus it is used for electro-optic and nonlinear optic applications [1, 2, 3, 4, 5, 6, 7, 8, 9, 10, 11]. It may say that refractive index (RI) is the most effective element in optical phenomena. Ledoux et al. first proposed a Sellmeier set for RI of NPP, based on measured refractive indices [3]. Banfi, Datta and co-workers design a more accurate Sellmeier set for RI of NPP for nonlinear optical studies [4, 5, 6]; these data based on classic and macroscopic measurements and approaches. Some authors explain RI in molecular bases [12, 13, 14], although no data on real material exist in this literature. Our purpose in this paper is the explanation of RI or dispersion effect of a real material, a single crystal film of NPP, in an approach

that name nano-quantum photonic [15, 16, 17]. This approach is based on four elements: 1- quasi-quantum principle for justification of phenomenon in molecular scales, 2- Knowledge of crystal network and its space shape, 3- Short range intramolecular and intermolecular forces, 4- Monte-Carlo time domain simulation. We suppose a laser beam is a flow of photons when passes through single crystal film, interacts with delocalization  $\pi$ -electron system of NPP molecule and delays the photon in every layer. By precise calculation of these retardation in every layer, we obtain refractive index (RI) in specific wavelength. The results obtained from this method are agreeable to experimental data. Our previous work was about MNA aromatic crystal, with a simpler structure than NPP molecule, [17, 18]. By simulation on more complex crystal, NPP, and considering the agreement of simulation results with experimental and Sellmeier data, the validity of assumptions and simulations will be proved. Hence, first we introduce crystal and molecular structure of NPP and comparison to Benzene molecule, after which we explain normal dispersion formula according to classical physics, electromagnetism, quantum electrodynamics and our microscopic model. In fourth section, we simulate and calculate the probability density function (PDF) of electron presence in benzene ring of NPP molecule, approximately. In fifth section, we express detailed calculation of RI. In sixth part, we clarify how to obtain RI for NPP crystal and compare simulation results with experimental and Sellmeier data.

## 2 Crystal and Molecular structure of NPP

Organic molecular units and conjugated polymer chains possessing  $\pi$ -electron systems usually form as centrosymmetric structures and thus, in the electric dipole approximation, would not show any nonlinear second order optical properties. The necessary acentric may be provided by first distorting the  $\pi$ -electron system by in-

\*Material presented in this paper is a part of Ali Akbar Wahedy Zarch's work on his thesis towards Ph.D degree. Dr. Hassan Kaatuzian is his advisor on thesis.



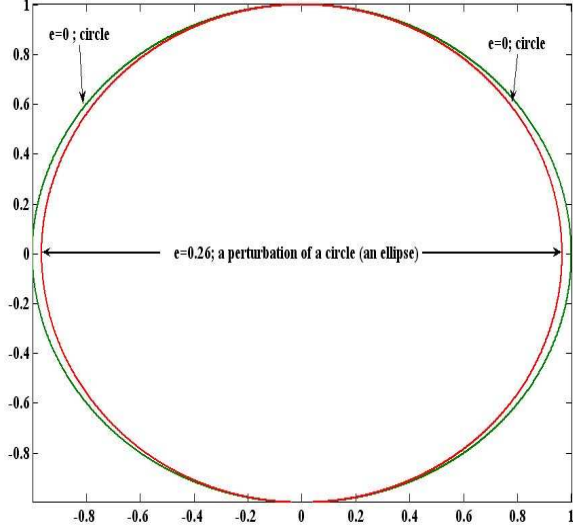


Figure 5: The comparison between circles for Benzene molecule electron cloud and NPP molecule electron cloud, (approximately).

### 3 Classic and microscopic model for dispersion

Cauchy suggested an equation that performs a relation between wavelength and RI in form [21]:

$$n - 1 = \frac{A}{\lambda^2} + \frac{B}{\lambda^4} + \frac{C}{\lambda^6} + \dots \quad (1)$$

If we have three RI in three wavelength according to experimental data, then we have RI in any wavelength in transparent range of that substance. Ledoux et.al [3] proposed a Sellmeier formula for NPP crystal in the form:

$$n^2 = A + \frac{B}{1 - \frac{C}{\lambda^2}} + D \cdot \lambda^2$$

and Datta et al. [5] demonstrated a more accurate of it in the form:

$$n^2 = A + \frac{B}{1 - \frac{C}{\lambda^2}} + \frac{D}{1 - \frac{E}{\lambda^2}}$$

that A,B,C,D and E is exerted from table 1. These formulas that derived from Sellmeier equation, gives no microscopic perspective of interactions of photon-electron when light passes through the material; additionally in sub-micron scales, RI loses its stabilization and these equations are not useful [15].

Electromagnetism have also a dispersion equation:

$$n - 1 = \frac{q_e^2}{2\epsilon_0 m} \sum_k \frac{N_k}{\omega_k^2 - \omega^2 + i\gamma_k \omega} \quad (2)$$

where there are  $N_k$  electrons per unit of volume, whose natural frequency is  $\omega_k$  and whose damping factor is  $\gamma_k$ , [8].

Quantum Electrodynamics attains a famous dispersion relation in form of:

$$n^2 - 1 = \frac{N}{\hbar\epsilon_0} \sum_{\nu} \left\{ \frac{(e_1 \cdot \mu_{1\nu})(e_1 \cdot \mu_{\nu 1})}{\omega_{\nu 1} + \omega} + \frac{(e_1 \cdot \mu_{1\nu})(e_1 \cdot \mu_{\nu 1})}{\omega_{\nu 1} - \omega} \right\} \quad (3)$$

by Density Matrix Method where:

$$W_n - W_m = \omega_{nm}$$

where W is the energy level and

$$\mu_{ik} = i \frac{e}{m\omega_{ik}} p_{ik}$$

that introduce the notation:

$$e_i = (e_x^{(i)}, e_y^{(i)}, e_z^{(i)}); \quad i = 1, 2$$

for unit of the polarization of the incident and scattered light, and

$$r = (x, y, z)$$

for position vector of the electron, and

$$p = (p_x, p_y, p_z)$$

for the electron momentum vector [22]. This formula is the simplest form in thermal equilibrium and is very complex in real world. This means that for attaining RI from quantum electrodynamics theory, we require to measure electron momentum vector and certainly, these elements can be calculated on the basis of semiclassical arguments.

Now, we suggest a microscopic model for RI and wavelength relationship. In this pattern, average photon flux is related to classic electromagnetic concepts like: intensity, power and energy. For example if we have a monochromatic laser beam with frequency  $\nu$  and intensity I, then we may attain average photon flux from relation [23]:

$$\phi = \frac{I}{h\nu} \quad (4)$$

now if we assume thin single crystal film of NPP in "b" direction of crystal (or "z" axis) radiated by a He-Ne laser with:  $\lambda=633$  nm, average power=10mw and beamwidth=20 microns, then from (4) average photon flux is equal to  $10^{22} \text{photons}/(\text{s} - \text{cm}^2)$  that signifies in every second  $10^{22}$  photons arrive to each centimeter square. Moreover from data of crystal in section (II) in every  $36.5(A^\circ^2)$  in z direction, one NPP molecule exists. Therefore in every second  $36.5 \times 10^6$  photons interact with any molecule or in other words in every 27ns, one photon interacts with any NPP molecules. In each interaction between photon and electron in every layer of crystal, we suppose delay time equal to  $\tau_i$  (ith layer of crystal). Total retardation time for m layers in crystal region is equal to:

$$\sum_{i=1}^m \tau_i$$

Table 1: Sellmeier data of NPP

Ledoux et al.[3] selmeier form					Datta and Banfi et al.[5] selmeier form				
$n$	$A$	$B$	$C$	$D$	$A$	$B$	$C$	$D$	$E$
$n_x$	2.3532	1.1299	0.1678	0.0392	2.41704	1.08674	0.16933	-0.34200	10
$n_y$	2.8137	0.3655	0.2030	-0.0816	2.76667	0.37156	0.20289	0.47880	10
$n_z$	2.1268	0.0527	0.1550	-0.0608	2.19965	0.00457	0.17160	0.59363	10

Consequently required time for photon transmission in L length of crystal is equal to  $\tau$ , achieved from relation:

$$\tau = \frac{L}{\frac{c_0}{n}} = \frac{nL}{c_0} = \frac{L}{c_0} + \sum_{i=1}^m \tau_i \quad (5)$$

or

$$n - 1 = \frac{c_0}{L} \sum_{i=1}^m \tau_i \quad (6)$$

where  $c_0$  is velocity of light in vacuum. By using this relation, we can relate macroscopic quantity  $n$  to microscopic quantity  $\tau_i$ . According to (3) if,  $n = 3, L = 3\mu$ , consequently  $\Sigma\tau$  is equal to  $10^{-14}$ sec. Because in b direction of crystal in  $3\mu$  length, approximately 4024 molecules exist, therefore the average quantity of  $\tau$ :

$$\bar{\tau} = \frac{\Sigma\tau}{N}$$

is in order of  $10^{-18}$  sec. From this quantity and previous quantity that in every 27ns, one photon interacts with one NPP molecule, we conclude that in every moment, just one photon interacts with one molecule. Because NPP molecule has delocalization electrons,(or  $\pi$ -electron system), in benzene ring, that photon interacts with this electron type and it is annihilated [24]. We call this photon, a successful photon, (that does not produce phonon). By this method, we are able to simulate microscopic perturbation of RI for NPP successfully. Equations (1), (2) and (3) use average quantities in frequency domain, but in (6), we utilize momentary quantities in time domain that perturb on average values. In our nano-quantum photonic model, we may estimate photon-electron mutual actions in very short time, on the order of "attosecond", ( $10^{-18}s$ ). We claim that our approach is applicable for optical material that their crystal structure is known, especially aromatic optical crystal. Because, they have a delocalization electron and successful photon interact with it. For inorganic optical material if we know active electron in optical phenomenon, we are able to use this method. In our approach, we do not have any measurement and we have simulation and estimation with an ideal observer. This is the first time that we have focused on a real material for approximating its constitutive parameters (like  $n, \epsilon, \sigma, \mu$ ). Neither in classical nor in Q.M. approach we only have typical material. In Q.E.D, we have not seen yet practical or predictable data for a real material. Our approach is quasi-classic and is similar to the Compton effect but in greater wavelengths. We have used some approximations for problem solution.

## 4 PDF calculation of $\pi$ -electron in benzene ring of NPP

To obtain  $\pi$ -electron wavefunction for benzene molecule the Schrödinger equation may be solved. Since this is very complex, this cannot be done exactly, an approximated procedure known as Hückle method must be employed. In this method, by using Hückle Molecular-Orbital (HMO) calculation, a wave function is formulated that is a linear combination of the atomic orbitals that have overlapped [20] (see Fig.3) (this method is often called the linear combination of atomic orbitals); that is:

$$\Psi = \sum_i C_i \Phi_i$$

where the  $\Phi_i$  refers to atomic orbitals of carbon atoms in the ring and the summation is over the six C atoms. The Schrödinger equation for a delocalized electron is:

$$\left[ \frac{-\hbar^2}{2m} \nabla^2 + \sum_i V_i \right] \Psi = E \Psi$$

that  $V_i$  is the atomic potential of  $i$ th atom. Following the common approach in quantum mechanics, we multiply the above equation from the left by  $\Phi_i$  and integrate over space. By solving this set of algebraic equations, the  $C_i$  coefficient, the  $\Phi_i$  wavefunction and energy E will be obtained, (Fig. 3). Thus, the  $|C_i|^2$  is the probability of the  $\pi$ -electron at  $i$ th atom. Thus:

$$|C_1|^2 + |C_2|^2 + |C_3|^2 + |C_4|^2 + |C_5|^2 + |C_6|^2 = 1$$

In the case of Benzene molecule:

$$|C_i|^2 = \frac{1}{6}$$

as followed from the symmetry of the ring [20, 25, 26]. But for NPP molecule there isn't such as Benzene molecule. NPP is a polar molecule. Nitro ( $NO_2$ ) is more powerful electronegative compound than prolinol and pulls  $\pi$ -electron system; consequently, the probability of finding  $\pi$ -electron system at various carbon atoms of main ring isn't same and the probability of finding  $\pi$ -electrons near the Nitro group is greater than near the prolinol group. Therefore there is no symmetry for NPP and electron cloud is spindly or oblong, (similar to dom-bell) (Fig. 6). We estimate this form of electron cloud by an ellipse that our calculations would be uncomplicated. According to Bohr atomic model, we assume  $\pi$ -electron is rotating around the center of positive charge in agreement with Kepler law. The positive



Figure 6: Assumed  $\pi$ -electron orbit of NPP molecule that obtained from fig. 5 for Benzene molecule (approximately).

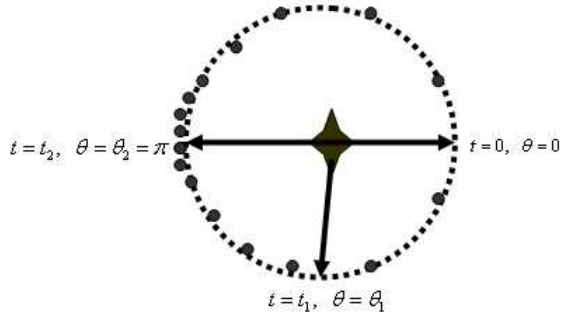


Figure 7: Bohr atomic model and Kepler law for  $\pi$ -electron system on an orbit.

charge in this approximation is virtual.

For attaining probability of electron presence on an orbit (fig. 7), we say,  $T$  time is required by radial vector to sweep total  $\pi.u.v$  interior area of ellipse ( $u$  and  $v$  are semimajor and semiminor axis of ellipse respectively), in  $t$  times, this radial vector sweeps

$$\pi.u.v.\frac{t}{T}$$

area of ellipse, (see fig. 7). If  $t$  is the time, that electron sweeps  $\theta$  radian of orbit then  $t$  is obtained from this relation [27]:

$$t = \frac{T}{2\pi} \left\{ 2 \arctan \left( \sqrt{\frac{1-\varepsilon}{1+\varepsilon}} \tan\left(\frac{\theta}{2}\right) \right) - \frac{\varepsilon \cdot \sqrt{1-\varepsilon^2} \cdot \sin(\theta)}{1+\varepsilon \cdot \cos(\theta)} \right\} \quad (7)$$

Where  $\varepsilon$  is ellipse eccentricity. By using this relation, we attain the required time for electron to traverse from  $\theta$  to  $\theta + d\theta$  ( $dt$ ) and it is divided by total time  $T$ . By this approach, we can determine the PDF approximately. Fig. 8 and Fig. 9 show PDF and CDF (Cumulative Density function) respectively for presence probability of  $\pi$ -electron around the assumed orbit in fig. 7. Such as shown in this figure the PDF in apogee (near the Nitro group), is maximum and in perigee (near the prolinol group) is minimum.

## 5 $\tau$ Calculation

The angle between  $Y$  vector and charge transfer action ( $N_1-N_2$ ) is  $58.6^\circ$  and  $X$  and  $Z$  axis is perpendicular to  $Y$  (Fig.10). We consider propagation along  $Z$  direction.

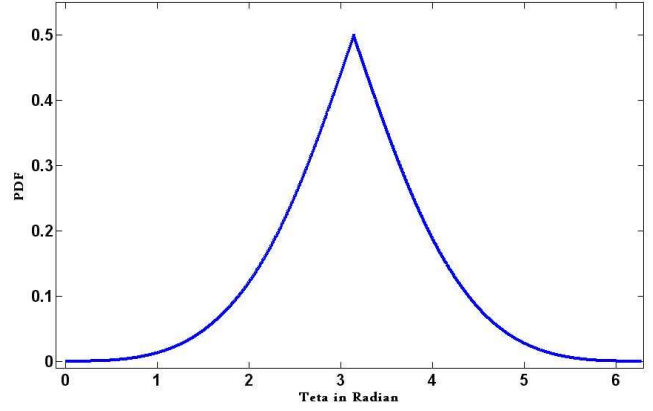


Figure 8: PDF of presence probability of  $\pi$ -electron around the assumed orbit in fig. 7;

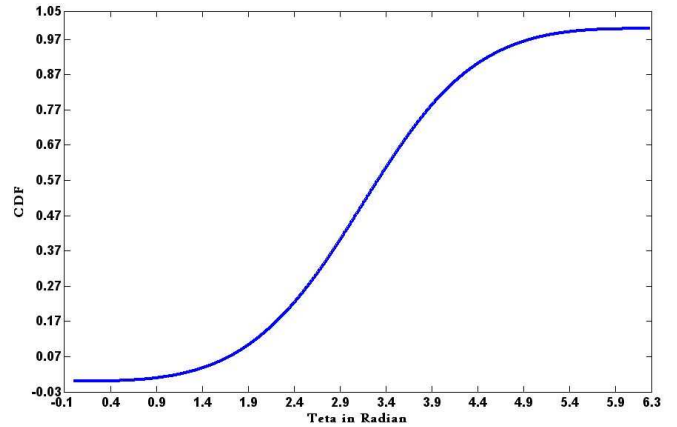


Figure 9: CDF of presence probability of  $\pi$ -electron around the assumed orbit in fig. 7;

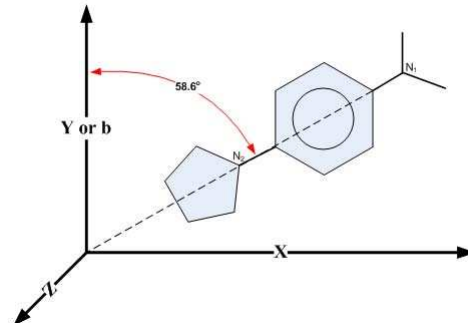


Figure 10:  $X$ ,  $Y$  and  $Z$  axis and NPP molecule in dielectric frame.

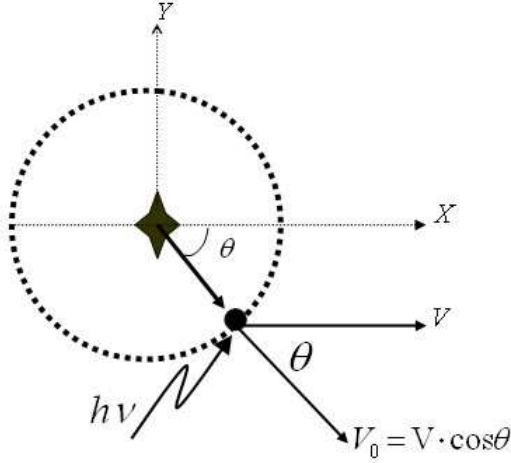


Figure 11: Calculation of  $\tau$

We spot a photon interacts with  $\pi$ -electron of NPP in first layer. After interaction, this photon gives its energy to electron and is annihilated. Electron absorbs energy and digresses in direction of photon momentum. Electron with photon energy, may not be unbounded and after arriving to apogee of digression, it returns back to main orbit, because the photon energy is equal to  $h\nu = 1.96\text{ev}$  ( $h$  is Planck's constant and  $\nu$  is frequency of laser beam) whereas energy for ionization is greater than  $5\text{ev}$ . When electron returns to main elliptical orbit one photon is produced. The time coming up and down is  $\tau$  delay time. This photon after freedom goes to second layer in direction of annihilated photon (nonce, we assume the polarization doesn't change), in second layer this photon interacts with another delocalization  $\pi$ -electron certainly, because the effective range of photon is approximately equal to its wavelength and is very greater than the distance between molecules. This action is repeated for each layer. The location of photon-electron interaction is significant in every molecule and it is effective on  $\tau$  quantity directly. We assume that interacting photon has a field in X direction and electron subject to virtual positive charge center. According to fig.11 we have:

$$h\nu = \frac{1}{2}mV^2 = \frac{1}{2}m\left(\frac{V_{ox}}{\cos(\theta)}\right)^2$$

and consequently:

$$V_{ox} = \sqrt{\frac{2h\nu}{m}} \cdot \cos(\theta)$$

that  $\frac{1}{2}mV^2$  is the kinetic energy of electron after interaction with photon, and columbic force,  $F$ , is equal to:

$$F = K \frac{Ze^2}{r^2} = ma$$

where  $e$ ,  $a$  and  $m$  are charge, acceleration and mass of electron respectively.  $Ze$  is positive charge equivalent

in effective center of positive charge. Consequently the time required for electron to go up and down is equal to:

$$\tau_x = \frac{V_{ox}}{a} = \frac{\sqrt{2h\nu \cdot m \cdot \cos(\theta)}}{KZe^2} \cdot r^2 \quad (8)$$

and similarly:

$$\tau_y = \frac{V_{oy}}{a} = \frac{\sqrt{2h\nu \cdot m \cdot \sin(\theta)}}{KZe^2} \cdot r^2 \quad (9)$$

that

$$r = \frac{(1 - \epsilon^2) \cdot u}{1 + \epsilon \cdot \cos(\theta)}$$

where  $\epsilon$  is the elliptical eccentricity and  $u$  is the semi-major axis of the ellipse. By performing  $\tau_x$  and  $\tau_y$  separately in every layer and using (6), we attain  $n_x$  and  $n_y$  separately.

## 6 Attaining of $n_x$ and $n_y$ for NPP crystal

We are now in a position to take up  $n_x$  and  $n_y$  by Monte-Carlo method, then we simulate random number producer using *MATLAB* program. This program produces PDF quantities explained in section (4) and relates each of them to every molecule. These values are indexing  $\pi$ -electron positions in each layer, by assumption a reference point (see fig. 7). Additionally we have used a *MATLAB* program for Monte-Carlo simulation. The inputs of this program are:

1. The wavelength of incident optical beam in which we want to obtain refractive index of NPP crystal;
2.  $h, m, q, k = \frac{1}{4\pi \cdot \epsilon}, c_0$  that are Planck's constant, electron rest mass, elementary charge, Coulomb constant and speed of light respectively.
3. Unit cell parameters of NPP crystal:  $a, b, c, \beta$  and its other parameters that have given in section 2.
4.  $L$ : crystal thickness that in our simulation it is  $3\mu\text{m}$ . And the outputs of *MATLAB* program are:  $n_x, n_y$  in each wavelength and their related errors from experimental data.

System calibration is done in this method that we obtain two refractive indexes in two wavelengths with  $\epsilon$  (eccentricity),  $u$  (semimajor axis of ellipse) and  $Z$  (equivalent positive charge) in a way that refractive indexes in two wavelengths are very close to experimental data. Then we would see that refractive index in other wavelengths with same  $\epsilon$ ,  $u$  and  $Z$  will be achieved. Of course these values,  $\epsilon$ ,  $u$  and  $Z$  would be close to experimental structure of crystal, for example  $u$  would be greater than and smaller than minimum and maximum sizes of six lengths of benzene hexagonal respectively, or  $\epsilon$  would be small and greater than zero. In other hand these values must be logical. From this method in our simulation we have obtained  $\epsilon = 0.26, Z = 3.9, u = 1.4A^0$  that is very close to experimental and structural data. Tables (2) and (3) compare the results of simulation with experimental data. From these tables we perceive  $n_x$  and

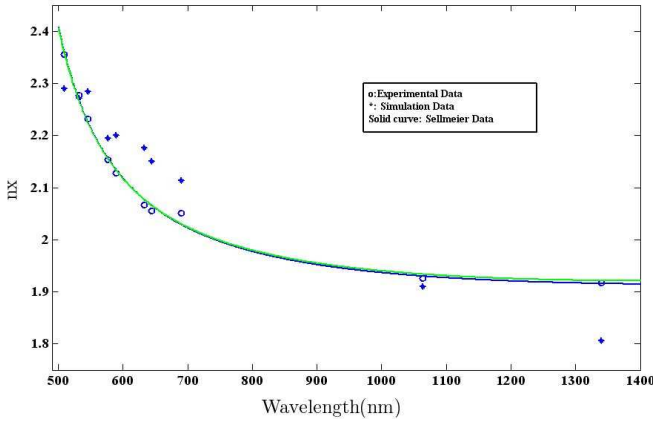
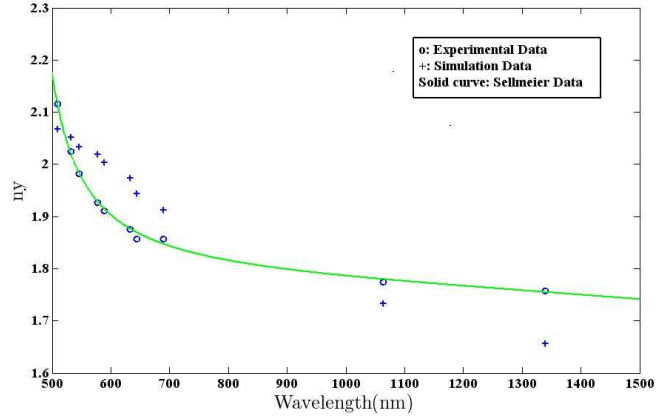


Table 2: Comparison of experimental data and simulation results for  $n_x$ 

Wavelength( $\lambda - nm$ )	509	532	546	577	589	633	644	690	1064	1340
$n_x(Experimental)[3]$	2.355	2.277	2.231	2.153	2.128	2.066	2.055	2.051	1.926	1.917
$n_x(Simulation)$	2.290	2.271	2.284	2.195	2.200	2.176	2.150	2.113	1.910	1.806
Error%	2.7	0.23	2.4	1.9	3.3	5.3	4.6	3	0.8	5.8

Table 3: Comparison of experimental data and simulation results for  $n_y$ 

Wavelength( $\lambda - nm$ )	509	532	546	577	589	633	644	690	1064	1340
$n_y(Experimental)[3]$	2.116	2.024	1.982	1.927	1.911	1.876	1.857	1.857	1.774	1.757
$n_y(Simulation)$	2.068	2.052	2.033	2.019	2.004	1.974	1.944	1.913	1.733	1.656
Error%	2.3	1.4	2.6	4.8	4.9	5.2	4.7	3	2.3	5.7

Figure 12: Comparison of experimental data, Simulation data and Sellmeier data for  $n_x$ Figure 13: Comparison of experimental data, Simulation data and Sellmeier data for  $n_y$ 

$n_y$  that we attained from simulation, have 5.8 percent maximum error and 0.23 percent minimum error. These values are acceptable measures. Figures 12 and 13 compare these data with Sellmeier data that are provided from [3, 5].

## 7 Conclusion

The suggested physical model in this paper that is based on assumption of energetic particles could be a powerful tool for analyzing and explaining processes that happen in waveguides in microscopic sizes. From similar models we can explain optical events such as second harmonic generation and optical polarization in fiber optic applications and optical communication. We will try to continue applying this method for justification of another phenomenon. Our approach is quasi-quantum and its advantages are:

- 1-We may to obtain scientific data of a real material,
- 2-We utilize momentary quantities in time domain,
- 3- Our method may predict some events in nanostructures and very short time by considering short-range interatomic forces and its effect on photons.

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